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Bir: ທີ່ Fransmitted herewith for filing is the patent application of

nventor(s):

Jashawant J. Modi

For:

Hydrophobically Modified Polysaccharides In Household Preparations

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	sheets of drawing.
	An Assignment of the Invention to Hercules Incorporated, Hercules Plaza, Wilmington, DE 19894-0001.
	A certified copy of a application.
X	A Combined Declaration and Power of Attorney.
	A verified statement to establish small entity status under 37 CFR 1.9 and 37 CFR 1.27.

X The filing fee has been calculated as shown below:

	(Col. 1)	(Col. 2)
FOR:	NO. FILE	NO. EXTRA
BASIC FEE		
TOTAL CLAIMS	33-20 =	13
INDEP CLAIMS	1-3=	
MULTIPLE DEPENDENT CLAIM PRESENTE		IM PRESENTED

RATE	FEE
	\$ 790.00
X 22 =	\$ 266.00
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+ 270 =	\$ 0
	\$1056 .00

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X Any patent application processing fees under 37 CFR 1.17.
The issue fee set in 37 CFR 1.18 at or before mailing of the Notice of Allowance, pursuant to 37 CFR 1.311(b).
X Any filing fees under 37 CFR 1.16 for presentation of extra claims.

Respectfully submitted,

David Edwards Registration No. 27,293

December 17 , 1997

b:PTO1082b.frm

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### HYDROPHOBICALLY MODIFIED POLYSACCHARIDES IN HOUSEHOLD PREPARATIONS

This invention relates to the use of hydrophobically modified polysaccharides in household care products (which are define as a cleaning, polishing, sanitizing, pesticidal, and toilet preparations). More specifically, this invention relates to the use of such polysaccharides in such products where the alkyl moiety of the hydrophobe has 3-24 carbon atoms.

#### BACKGROUND OF THE INVENTION

Prior to the present invention, water soluble polysaccharides have been used in cleaning, sanitizing, polishing, toilet preparations, and pesticide preparations; applications such as air deodorants/fresheners, rug and upholstery shampoos, insect repellent lotions, all purpose kitchen cleaner and disinfectants, toilet bowl cleaners, fabric softener-detergent combinations, fabric softeners, fabric sizing agents, dishwashing detergents, vehicle cleaners and shampoos. Widely used commercially available polysaccharides include water soluble polysaccharide ethers such as methyl cellulose (MC), hydroxypropylmethylcellulose (HPMC), hydroxypthylcellulose (HEC), hydroxypropyl (HP) guar, hydroxypthyl guar, guar, starch, and other nonionic starch and guar derivatives. The use of these prior art polysaccharides in such products are sometimes associated with processing difficulties such as compatibility with other ingredients, solubility with certain other ingredients, solution clarity (when needed) and stability under alkaline (or acidic) conditions of the products.

US Patent numbers 5,106,609, 5,104,646, and 5,100,658 are examples of patents that disclose the use of hydrophobically modified cellulose ethers in cosmetic products. These patents disclose the use of high molecular weight (i.e., 300,000 to 700,000) and long chain alkyl carbon substitution in the hydrophobe (i.e., 8 to 24 carbons) for use in cosmetic composition. Also, US

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derivatives, modified with long chain alkyl group substitution in the hydrophobe. US Patent number 4,845,207 discloses a hydrophobically modified nonionic, water-soluble cellulose ether and US Patent 4,939,192 discloses the use of such ether in building compositions.

Certain of the prior art cellulose ethers have poor compatibility with salts or poor solubility in certain solvents such as polyhydric alcohols used in cleaning, sanitizing, polishing, pesticide and toilet preparation applications while others are not tolerant of alkaline or acidic conditions. Hence, a need still exists in the industry to have cellulose ethers that have good compatibility with salts, good solubility in certain solvents, and tolerance to acidic or alkaline conditions while producing products that do not have color problems, when desired.

#### SUMMARY OF THE INVENTION

The present invention is directed to a household product (which is defined as a cleaning, sanitizing, polishing, pesticide or toilet preparation) composition comprising:

- (a) from about 0.1% to about 99% by weight of a vehicle system which comprises a hydrophobically modified water soluble polysaccharide polymer which comprises a water soluble polysaccharide polymer backbone, a hydrophobic moiety selected from the group consisting of:

  1) 3-alkoxy-2-hydroxypropyl group wherein the alkyl moiety is a straight or branch chain having 3-24 carbon atoms, or 2) C<sub>3</sub>-C<sub>24</sub> alkyl, aryl alkyl, alkyl aryl groups and mixtures thereof, wherein the hydrophobic moiety is present in an amount up to the amount which renders said polysaccharide less than 1% by weight soluble in water, and
- (b) at least one other cleaning, sanitizing, polishing, pesticide or toilet preparation product ingredient.

#### DETAILED DESCRIPTION OF THE INVENTION

It has been surprisingly found that hydrophobically modified polysaccharides have various advantageous properties over prior art water soluble polysaccharides in cleaning, polishing, sanitizing, pesticidal, toilet or perfume products. Any water soluble polysaccharide or derivatives can be used as the backbone to form the hydrophobically modified polysaccharide of this invention. Thus, e.g., hydroxyethylcellulose (HEC), hydroxypropylcellulose (HPC),

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methylcellulose (MC), hydroxypropylmethylcellulose (HPMC), ethylhydroxyethylcellulose (EHEC), and methylhydroxyethylcellulose (MHEC) and, agar, dextran, locust bean gum, starch, guar and their nonionic derivatives can all be modified. The amount of nonionic substituent such as methyl, hydroxyethyl, or hydroxypropyl does not appear to be critical so long as there is a sufficient amount to assure that the ether is water soluble. The polysaccharides of this invention have a sufficient degree of nonionic substitution to cause them to be water soluble and a hydrophobic moiety where selected from the group consisting of 1) 3-alkoxy-2-hydroxypropyl group wherein the alkyl moiety is a straight or branch chain having 3-24 carbon atoms, or 2) C<sub>3</sub>-C<sub>24</sub> alkyl, aryl alkyl, alkyl aryl groups and mixtures thereof, wherein the hydrophobic moiety is present in an amount up to the amount which renders said polysaccharide less than 1% by weight soluble in water. When the hydrophobe is an alkyl, aryl alkyl, or alkyl aryl moiety, the number of carbons can be 3-24, preferably 3-22, more preferably 4-18, and most preferably 4-16.

The preferred polysaccharide backbone is hydroxyethylcellulose (HEC). The HEC which is modified to function in this invention is a commercially available material. Suitable commercially available materials are marketed by the Aqualon Company, a division of Hercules Incorporated, Wilmington, Delaware U.S.A., under the trademark Natrosol<sup>®</sup>.

The alkyl modifier, cationic group, anionic group and zwitterionic group can be attached to the polysaccharide backbone via an ether, ester, or urethane linkage. Ether is the preferred linkage as the reagents most commonly used to effect etherification because it is readily obtainable; the reaction is similar to that commonly used for the initial etherification, and the reagents used in the reaction are usually more easily handled than the reagents used for modification via the other linkages. The resulting linkage is also usually more resistant to further reactions.

An example of the polysaccharide of the present invention is the 3-alkoxy-2-hydroxypropylhydroxyethylcellulose that is substantially completely soluble in water at ambient temperature.

The hydrophobic moiety is generally contained in an amount of about 0.05 to about 50 wt. %, preferably about 0.1 to about 25 wt. %, based on the dry weight of the substituted polymer. The alkyl group of the 3-alkoxy-2-hydroxypropyl group can be a straight chain alkyl group or

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branched alkyl group having 3 to 24 carbon atoms. Exemplary modifying radicals are propyl-, butyl-, pentyl-, 2-ethylhexyl, octyl, cetyl, octadecyl, and docosapolyenoic glycidyl ether.

The hydrophobically modified polysaccharide of the present invention is an essential ingredient of the system. Another ingredient that may be in the system is a surfactant that can be either soluble or insoluble in the composition. A compatible solvent may also be used in the system that can be either a single solvent or a blend of solvents.

Examples of the surfactants are anionic, nonionic, cationic, zwitterionic, or amphoteric type of surfactants. The surfactant can be soluble or insoluble in the present invention and (when used) is present in the composition of from 0.01 to about 50% by weight of the composition. Synthetic anionic surfactants include alkyl and alkyl ether sulfates.

Specific examples of alkyl ether sulfates which can be used in the present invention are sodium coconut alkyl trimethylene glycol ether sulfate; sodium tallow alkyl trimethylene glycol ether sulfate; and sodium tallow alkyl hexaoxyethylene sulfate. Sodium tallow alkyl diethylene glycol ether sulfate; and sodium tallow alkyl sulfate.

Nonionic surfactants, can be broadly defined as compounds containing a hydrophobic moiety and a nonionic hydrophilic moiety. Examples of the hydrophobic moiety can be alkyl, alkyl aromatic, dialkyl siloxane, polyoxyalkylene, and fluoro-substituted alkyls. Examples of hydrophilic moieties are polyoxyalkylenes, phosphine oxides, sulfoxides, amine oxides, and amides.

Cationic surfactants useful in vehicle systems of the compositions of the present invention, contain amino or quaternary ammonium hydrophilic moieties which are positively charged when dissolved in the aqueous composition of the present invention.

Zwitterionic surfactants are exemplified by those which can be broadly described as derivative of aliphatic quaternary ammonium, phosphonium, and sulfonium compounds, which can be broadly described as derivative of aliphatic quaternary ammonium, phosphonium, and sulfonium compounds, in which the aliphatic radicals can be straight or branched chain, and wherein one of the aliphatic substituents contains from about 8 to about 18 carbon atoms and one contains as anionic water-solubilizing group, e.g., carboxy, sulfonate, sulfate, phosphate, or phosphonate.

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Examples of amphoteric surfactants which can be used in the vehicle systems of the compositions of the present invention are those which are broadly described as derivatives of aliphatic secondary and tertiary amines in which the aliphatic radical can be straight or branched chain and wherein one of the aliphatic substituents contains from about 8 to about 18 carbon atoms and one contains an anionic water solubilizing group, e.g., carboxy, sulfonate, sulfate, phosphate, or phosphonate.

According to the present invention, the solvent used in the system should be compatible with the other components in the present composition. Examples of the solvents used in the present invention are water, water-lower alkanols mixtures, and polyhydric alcohols having from 3 to 6 carbon atoms and from 2 to 6 hydroxyl groups. Preferred solvents are water, propylene glycol, water-glycerine, sorbitol-water, and water-ethanol. The solvent (when used) in the present invention is present in the composition at a level of from 0.1% to 99% by weight of the composition.

The active component is optional because the dissolved polymer can be the active ingredient component. An example of this is the use of the polymer in a fabric sizing spray. However, when an active ingredient is needed, it must provide some benefit to the user. Examples of substances that may suitably be included according to the present invention are as follows:

- 1) Perfumes, which give rise to an olfactory response in the form of a fragrance and deodorant perfumes which in addition to providing a fragrance response can also reduce odor;
- 2) Insect repellent agent whose function is to keep insects from a particular area or attacking skin;
  - 3) Bubble generating agent, such as surfactants which generates foam or lather;
  - 4) Pet deodorizer such as pyrethrins which reduces pet odor;

- 5) Pet shampoo actives, whose function is to remove dirt, foreign material and germs from the skin and hair surfaces;
- 6) Industrial grade bar and liquid soap actives that remove germs, dirt, grease and oil from skin and sensitizes skin;
- 7) All purpose cleaning agents, that remove dirt, oil, grease, germs from the surface in area such as kitchens, bathroom, public facilities
- 8) Disinfecting ingredients that kill or prevent growth of germs in a house or public facility;
- 9) Rug and Upholstery cleaning actives which lift and remove dirt and foreign particles from the surfaces and also deliver softening and perfumes;
  - 10) Laundry softener actives which reduces static and makes fabric feel softer;
  - 11) Laundry detergent ingredients which remove dirt, oil, grease, stains and kills germs;
  - 12) Dishwashing detergent actives which removes stains, food, germs;
  - 13) Toilet bowl cleaning agents which removes stains, kills germs, and deodorizes;
  - 14) Laundry prespotter actives which helps in removing stains from clothes;
  - 15) Fabric sizing agent which enhances appearance of the fabric;
  - 16) Vehicle cleaning actives which removes dirt, grease, etc. from vehicles and equipments.

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## 17) Lubricating agent which reduces friction between parts.

The above list is only examples and is not a complete list of active ingredients that can be used. Other ingredients that are use in these types of products are well known in the industry. In addition to the above ingredients conventionally used, the composition according to the present invention can optionally also include ingredients such as a colorant, preservative, antioxidant, activity enhancer, emulsifiers, viscositying agents (such as salts, i.e., NaCl, NH<sub>4</sub>Cl & KCl), alcohol and fats and oils.

The vehicle systems and compositions of the present invention can be made using conventional formulation and mixing techniques. Methods of making various types of compositions are described more specifically in the following examples. The following examples are merely set forth for illustrative purpose, but it to be understood that other modifications of the present invention within the skill of artisans in the industry can be made without departing from the spirit and scope of the invention.

## Example 1 Toilet Soap Formula

	<u>Ingredients</u>	Weight %
	A. Water	65.70
	B. Sodium C14-C16 olefin sulfonate	20.00
	C. Sodium lauroyl sarcosinate	10.00
5	D. Cocamide monoethanol amine	3.00
	E. Natrosol-Plus®* 330	0.75
	F. Natrosol 250HR**	0.25
	G. Disodium EDTA (Disodium ethylene diamine tetra acetate)	0.20
	H. Methylparaben	0.10
		100.00

#### 10 Procedure

- 1. The Natrosol-Plus and Natrosol 250HR products were dispersed in water and the pH was raised to about 8.0 8.5 while stirring for 45 minutes to dissolve the polymer mixture to form a solution. Methylparaben was then added to this solution.
- 2. In a separate vessel, the surfactants (components B, C, & D) were combined, heated to 80°C, and mixed until homogeneous.
- 3. The surfactant solution was then added to the water-soluble polymer solution and mixed until well blended.
- 4. The Disodium EDTA was added to the blended solution and mixed for about 15 minutes, and then cooled to room temperature.

<sup>\*</sup>This polymer is hydrophobically modified hydroxyethylcellulose that is nonionic and contains both hydroxyethyl and long chain (i.e., C<sub>16</sub>) alkyl group; and has Brookfield viscosity of 150-750 cps at 1% at 25°C.

<sup>\*\*</sup>This product is hydroxyethylcellulose that is nonionic and has a Brookfield viscosity of 1500-2500 at 1% at 25°C.

## Source and Description of Products Used

	Generic or <u>CTFA Adopted Name</u>	<u>Trademark</u>	<u>Supplier</u>
5	Sodium C14-C16 olefin sulfonate	Bio-Terge AS-40	Stepan Chemical Company Northfield, Illinois
	Sodium lauroyl sarcosinate	Hamposyl L-30	W. R. Grace & Company Nashua, New Hampshire
10	Cocamide MEA	Monamid CMA	Mona Industries Inc. Paterson, New Jersey
		Natrosol 25HR Natrosol-Plus 330	Hercules Incorporated Wilmington, Delaware Hercules Incorporated Wilmington, Delaware

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## Example 2 Air Freshener Formulations - Hot Process

	<u>Ingredients</u>	Weight %
Α	Carrageenan (CHP1** from Hercules Incorporated)	02.00%
В	Locust bean Gum	00.25%
C	HMHEC 1* (Hercules Incorporated)	00.75%
D	Water	93.00%
Е	Propylene glycol (humectant, solubilizer)	02.00%
F	Germaben II*** (Sutton Laboratory, Chatham, NJ)	01.00%
G	Fragrance**** (Cinnamon)	<u>01.00%</u>
	Total	100.00%

#### **Procedure**

Water was heated to about 90°C while being stirred and polymers A, B, and C were added to the vortex of water and mixed for 10 minutes or until dissolved. Next, the propylene glycol was added to the solution and mixed for another 10 minutes and then cooled to 70°C while stirring. The preservative Germaben II was added and mixed for five minutes. The fragrance was added and mixed for five minutes and the mixture was transferred to a pack out container and cooled to room temperature.

<sup>\*</sup> This compound HMHEC 1 is 3-butoxy-2-hydroxypropylhydroxyethylcellulose that has an aqueous viscosity at 25°C of a minimum of 2500 cps at 1%, measured on a Brookfield LVTD Viscometer, and a cloud point of about 72°-78°C, that is treated with glyoxal

<sup>\*\*</sup> CHP1: Kappa carrageenan, not standardized with sugar or salt.

<sup>\*\*\*</sup> Germaben II: Preservative, Diazolidinyl urea.

<sup>\*\*\*\*</sup> Fragrance: Cinnamon, Flavor and Fragrance Specialties - Franklin Lake, NJ.

## Example 3 Air Freshener Formulations - Hot Process

	<u>Ingredients</u>	Weight %
A	Carrageenan (Genugel RLV* from Hercules Incorporated)	01.13%
В	HMHEC 1 (Hercules Incorporated)	00.50%
C	Fragrance (Cinnamon)	30.00%
D	Water	67.12%
E	Surfactant (Dowfax 2A1**, Dow Chemicals, Midland, MI)	00.25%
F	Germaben II (Sutton Laboratory)	01.00%
	Total	100.00%

#### **Procedure**

Water was heated to about 90°C while being stirred and polymers A and B were added to the vortex of water and mixed for 10 minutes or until dissolved and then cooled to 80°C while continued stirring. Next, the preservative Germaben II and surfactant were added and mixed for five minutes. The fragrance was added and mixed for five minutes and the hot mixture was injected into a porous foam substrate and cooled to room temperature and sealed into a nonpermeable wrap.

<sup>\*</sup> Genugel RLV: Carrageenan standardized with sugar

<sup>\*\*</sup> Dowfax 2A1: Benzene, 1,1-oxybis, tetrapropylene derivative, sulfonated sodium from Dow Chemical.

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## Example 4 Air Freshener Formulations - Hot Process

	<u>Ingredients</u>	Weight %
Α	Agar	01.00%
В	Starch	00.25%
C	Natrosol-Plus 430* (Hercules Incorporated)	00.25%
D	Water	96.50%
E	Germaben II (Sutton Laboratory)	01.00%
F	Fragrance (Cinnamon)	<u>01.00%</u>
	Total	100.00%

## **Procedure**

Water was heated to about 90°C while being stirred and polymers A, B, and C were added to the vortex of water and mixed for 10 minutes or until dissolved. Next, the solution was cooled to 70°C while stirring. The preservative Germaben II was added and mixed for five minutes. The fragrance was added and mixed for five minutes and the mixture was transferred to a pack out container and cooled to room temperature.

<sup>\*</sup>Natrosol-Plus 430 is nonionic hydrophobically modified hydroxyethylcellulose. It has long chain (C<sub>16</sub>) alkyl group. Aqueous viscosity at 1.0% is between 5000 - 9000 cps Brookfield viscometer at spindle 3, 6 rpm.

## Example 5 Air Freshener Formulations - Cold Process

	<b>Phase</b>	<u>Ingredients</u>	Weight %
	A	DASC (dihydroxyaluminum sodium carbonate)	00.30%
5	В	CMC 7MT** (Hercules Incorporated)	03.00%
	C	HMHEC 2*	00.25%
	D	Potassium Sorbate	00.12%
	E	Propylene glycol	05.00%
	F	Water	89.33%
10	G	Fumeric Acid	01.00%
	H	Fragrance***	01.00%
		Total	100.00%

#### **Procedure**

DASC and potassium sorbate were added to the vortex of water in a container while stirring. In a separate container, ingredients B, C, and G (CMC, HMHEC 2 and fumeric acid) were preblended and then the propylene glycol was slurried into the preblend. The slurry was immediately added to the DASC dispersion and mixed for 15 minutes. Next, the fragrance was added and mixed for five minutes or until well dispersed and the formulation was filled into a container and capped.

<sup>\*</sup> HMHEC 2 is 3-butoxy-2-hydroxypropylhydroxyethylcellulose that has aqueous viscosity at 25°C of a minimum of 2000 cps at 1% measured on a Brookfield LVTD Viscometer and a cloud point of about 62°-68°C without glyoxal treatment.

<sup>\*\*</sup> CMC 7MT: Sodium carboxymethylcellulose with 0.65 - 0.90

<sup>\*\*\*</sup> Fragrance: Wildflower from Flavor and Fragrance Specialties. degree of carboxymethyl substitution and 2% aqueous viscosity of 300-600 eps at 25°C.

# Example 6 Air Freshener Formulations - Cold Process

	Phase I	<u>Ingredients</u>	Weight %
	Α	CMC 7MF* (Hercules Inc.)	03.10%
5	В	HMHEC 2	00.20%
Ü	C	Propylene glycol	01.00%
	D	Water	<u>95.70%</u>
	_	Total	100.00%

#### **Procedure**

Propylene glycol was added to the vortex of water in a vessel and mixed for 5 minutes.

Polymers A & B were added to the vortex of the water/propylene glycol mixture and mixed for 20 minutes or until dissolved.

Phase II	<u>Ingredients</u>	Weight %
Α	(AAB) (Aluminum Acetate Basic pure, Aldrich, Cat #28982-5)	02.50%
В	Water	<u>97.50%</u>
	Total	100.00%

### **Procedure**

In a separate vessel A was added to water.

To 80 grams of Phase I, 20.0 grams of Phase II were added to the vortex of Phase I and mixed for 5 minutes. The product was poured into a container and allowed to be crosslinked. The gel strength can be increased by increasing the concentration of Phase I and/or Phase II.

<sup>\*</sup> CMC 7MF: Sodium carboxymethyl cellulose with 0.65 - 0.90 degree of carboxymethyl substitution and 2% aqueous viscosity of 400-800 cps at 25°C.

## Example 7 Insect Repellent Formulations

	<u>Ingredients</u>	Weight %
Α	Carrageenan (CHP1 from Hercules Incorporated)	02.00%
В	CMC 7LT* (Hercules Incorporated)	00.50%
C	HMHEC 2 (Hercules Incorporated)	00.75%
D	Water	92.75%
E	Propylene glycol (humectant, solubilizer)	02.00%
F	Germaben II (Sutton Laboratory)	01.00%
G	Insect Repellent (Flavor and Fragrance, Franklin Lakes, NJ)	01.00%
	Total	100.00%

### **Procedure**

Water was heated to about 90°C in a container and polymers A, B & C were added to the vortex of water while stirring and mixed for 10 minutes or until dissolved. Propylene glycol was added next and mixed for 5 minutes. The mixture was cooled to 70°C while mixing. Preservative Germaben II was added and mixed for five minutes. Insect repellant was then added and mixed for five minutes. This formulation was then transferred to a pack out container and cooled to room temperature.

<sup>\*</sup> CMC 7LT: Sodium carboxymethyl cellulose with 0.65 - 0.90 degree of carboxymethyl substitution and 2% aqueous viscosity of 25-50 cps at 25°C.

# Example 8 Pet Shampoo

		<u>Ingredients</u>	Weight %
	Α	Distilled water	69.70%
	В	Triethanol amine-lauryl sulfate (40% active)	15.00%
5	Č	Sodium lauroamphoteric (and) sodium trideceth sulfate	10.00%
3	Ď	Cocoamide diethanol amine	02.50%
	E	Glycol stearate	01.20%
	F	Propylene glycol(and) diazolidinyl urea (and)	00.75%
	•	Methyl paraben (and) propylparaben. (Preservative)	
	G	HMHEC 1	00.60%
10	H	Fragrance (Wildflower)	00.25%
10	I	Citric acid	<u>pH adjust</u>
MANTEN .	<del></del>	Total	100.00%

### **Procedure**

Polymer G was added to vortex of water in a vessel while being heated to 70°C and stirred for five minutes. Next, TEAL and glycol stearate were added to the vessel in small quantities and mixed well between the additions. After all of the additions were made, the heat was turned off and the vessel was allowed to cool. When the vessel was cooled down to about 55°C, Cocamide DEA was added to the vessel. Next, the preservative was added. The pH in the vessel then was adjusted to about 5.0 with citric acid solution. The fragrance was added and mixed for five minutes. The formulation was then poured into a pack out container and the top of the container was fastened.

# Example 9 Pet Shampoo

		<u>Ingredients</u>	Weight %
	Α	Distilled water	69.45%
		Triethanol amine-lauryl sulfate (40% active)	15.00%
	В	I Thethanol allime-laury surface (40% delive)	10.00%
5	C	Sodium lauroamphoteric (and) sodium trideceth sulfate	
	D	Cocoamide diethanol amine	02.50%
	_		01.20%
	E	Glycol stearate	00.75%
	$\mathbf{F}$	Propylene glycol(and) diazolidinyl urea (and)	00.7370
		Methyl paraben (and) propylparaben (Preservative)	
	~	HMHEC 1	00.60%
10	G		00.50%
	$\mathbf{H}$	Pyrethrins (10%)	
	Т	Citric acid	<u>pH adjust</u>
	1	Total	100.00%

## **Procedure**

Water was heated to 70°C in a vessel while being stirred and the polymer was added to the vortex of the stirred water mixed for five minutes. Next, TEAL and glycol stearate were added in small quantities at a time to the vessel and mixed well between each addition. The heat was then turned off and the vessel was allowed to cool. After the vessel cooled to about 55°C, Cocamide DEA were added to the vessel. Next, the preservative was added. Then, the pH of the vessel was adjusted to about 5.0 with citric acid solution. Pyrethrin was added and mixed for an additional five minutes. The formulation was then poured into pack out containers.

# Example 10 Insect Repellent Lotion

	<b>Phase</b>	<u>Ingredients</u>	Weight %
	A	HMHEC1	00.50%
		Distilled water	78.25%
		Glycerin, USP	02.00%
5	В	Glycol stearate	02.75%
		Stearic acid	02.50%
		Mineral oil	02.00%
		Acetylated lanolin	00.50%
		Cetyl alcohol	00.25%
	С	Distilled water	09.00%
		Triethanolamine	00.50%
in the second se	D	Insect Repellent	01.00%
2		(Flavor and Fragrance, Franklin Lakes, NJ)	00.50%
or it. Been then been been been been been been been b		Propylene glycol and diazolidinyl urea and Methylparaben and propylparaben (Germaben II - Sutton Lab, Chatham, NJ)	00.75%
		Total	100.00%

#### **Procedure**

HMHEC1 was dispersed in a vessel of well-agitated water (of Part A) by adding it to the vortex of the water. Next, glycerin was added to the vessel of water and polymer while mixing pH was adjusted to 8.5, and heated to 80°C. This slurry was mixed until dissolved. In a separate vessel, the Part B ingredients were blended together while heating to 80°C. This was mixed until dissolved. Part A was added to Part B with good agitation while the emulsion temperature of 80°C was maintained. Next, Part C ingredients were combined in a separate vessel and added to the emulsion and mixed continuously while cooling to 40°C. Then, Part D ingredients including the insect repellant were added to the emulsion and mixed for five minutes. The preservative was added to the emulsion and mixed for 5 minutes or until mixed well. The formulation was then cooled to room temperature and filled into containers.

# Example 11 Institutional Liquid Soap

	<b>Ingredients</b>	Weight %
	Water	74.88%
5	Sodium C14-C16 olefin sulfonate, 40% active	07.50%
	Sodium lauroyl sarcosinate, 30% active	06.66%
	Cocamidopropyl betaine, 35% active	06.66%
	Glycol stearate	01.00%
	HMHEC 1	00.80%
10	Propylene glycol	00.50%
10		00.50%
	Glycerin Tetrasodium ethylene diamine tetracetate	00.30%
	Stearalkonium chloride	00.10%
	· · ·	01.00%
_	Chlorhexidine digluconate (20%)	00.10%
15	Methylparaben	100.00%
tenderste Antonio	Total	100.0070

### **Procedure**

HMHEC 1 was dispersed in a container of stirred water and the pH was adjusted to about 8.0 - 8.5 while mixing to dissolve the polymer. The dissolution took about 45 minutes. The methylparaben was then added to the solution. While slowly stirring the water-soluble polymer solution, stearalkonium chloride, olefin sulfonate, and glycol stearate were added one at a time to the solution while stirring for five minutes between each addition. Next, the mixture was heated to 80°C until all of the glycol stearate was dissolved and the solution turned opaque. Next, the remaining ingredients were added while cooling the solution slowly to room temperature. The color, cyclohexidine, and fragrance ingredients were added to complete the formulation. Then, the formulation was packaged.

## Example 12 All-Purpose Cleaner

	<u>Ingredients</u>	Weight %
5	HMHEC 3* Rhodapex CD-128 (ammonium capryleth sulfate - Rhone-Poulenc) Butyl Cellosolve Sodium Metasilicate Trisodium Phosphate	00.30% 01.70% 03.50% 01.70% 01.00%
10	Water	91.80% 100.00%

### **Procedure**

The polymer (HMHEC 3) was added to the vortex of stirred water in a vessel and the pH of the vessel was adjusted to 8.5 while mixing for 45 minutes or until fully dissolved. All of the other ingredients were then added to the vessel in the order they are listed above. Between the addition of each ingredient to the solution in the vessel, the solution was stirred for five minutes.

For aerosol use, 85% of the solution was mixed with 15% propellant Genetron 12, from Allied Corporation.

<sup>\*</sup>HMHEC 3 is 3-butoxy-2-hydroxypropylhydroxyethylcellulose that has aqueous viscosity at 25°C of a minimum of 2000 cps at 1% measured on a Brookfield LVTD Viscometer and a cloud point of about 62°-68°C with glyoxal treatment.

## Example 13 Disinfectant

	<u>Ingredients</u>	Weight %
Α	Water	90.75%
В	HMHEC 3	00.75%
C	Benzalkonium	05.00%
	(Barquat 80-28 from Lonza Inc. Fair Lawn NJ)	
D	Nonionic surfactant	01.00%
E	Isopropanol	02.00%
F	Lemon Oil	00.50%
	Total	100.00%

## **Procedure**

The polymer (HMHEC 3) was added to the vortex of stirred water in a vessel and the pH of the vessel was adjusted to 8.5 while mixing for 45 minutes or until dissolved. All of the other ingredients were then added to the vessel in the order they are listed above. Between the addition of each ingredient to the solution in the vessel, the solution was stirred for about five minutes or until the ingredient was well mixed into the solution. The formulation was then poured into a pack out container.

## Example 14 Soap Bar

	Phase I	<b>Ingredients</b>	Weight %
	Α	Sodium cocoylisethionate	70.00%
	7.	(64% active, Geropon AS200 from Rhone Poulenc)	15.30%
5	В	Stearic acid, triple pressed	05.00%
J	Č	Hectorite	01.00%
	D	HMHEC 3	
	Phase II		
	Α	Water	07.49%
10	В	Sodium chloride	00.50%
10	Č	Dye (FD&C Red No. 40)	00.01%
	D	Titanium dioxide	00.20%
	Phase III		
TOTAL CONTROL OF THE	Α	Fragrance	00.50%
Control of the contro	Α	Total	100.00%
2 000		<del> </del>	

### **Procedure**

First, all the ingredients in Phase I were dry blended uniformly using a kitchen mixer. In a separate container, the Phase II ingredients, NaCl, titanium dioxide and dye, were mixed with water and then added to the Phase I ingredients in the kitchen mixer and continued to be mixed. Phase III ingredient was then added to the mixer containing the Phase I & II ingredients while mixing and the mixing was continued until the ingredients in the mixer were homogeneous. This formulation was then extruded through an extruder and packaged.

# Example 15 Rug and Upholstery Shampoo

	<b>Ingredients</b>	Weight %
	HMHEC 1	00.50%
5	Rhodapex ES (sodium laureth sulfate -	
•	Rhone-Poulenc)	20.00%
	Sodium tripolyphosphate	02.00%
	Ethyl carbitol	01.50%
	Tinopal 5BM (Triazinyl stilbene - Ciba-	
10	Geigy Corp.)	00.05%
	Water	<u>75.95%</u>
	Total	100.00%

## **Procedure**

The polymer HMHEC 1 was added to the vortex of stirred water in a vessel and the pH of the mixture in the vessel was adjusted to about 8.5 and stirred for about 45 minutes or until dissolved. The other ingredients were added one at a time in the order listed above. Each ingredient was mixed into the solution for 5 minutes at slow speed. After all of the additions, the formulation was poured into a container.

# Example 16 Liquid Laundry Softener-Detergent

	<u>Ingredients</u>	Weight %
	Water deionized	46.70%
5	HMHEC 3	00.50%
3	Varisoft 3690 (Imidazolium compound - Witco)	07.00%
	Varonic T210 (PEG-10 hydrogenated	25.00%
	tallow amine - Witco)	05.00%
10	Ethyl alcohol Citric Acid (15%)	15.80%
	Total	100.00%

### **Procedure**

The polymer HMHEC 3 was added to the vortex of stirred water in a vessel, the pH was adjusted to about 8.5, and was heated to about 60°C and mixed for about 30 minutes or until the polymer was dissolved. Varonic ingredient was added to the vessel and mixed until fully dissolved. Next, the Varisoft ingredient was added slowly while mixing. Heat was then turned off and mixing was continued in the vessel until the solution in the vessel was cooled. The remaining ingredients were added to the vessel and mixed. During the mixing cycle the rate of agitation was adjusted to minimize foaming. The formulation was then poured into a container.

# Example 17 Bacteriostatic Laundry Softener

<b>Ingredients</b>	Weight %
Water	06.20%
HMHEC 3	00.50%
Bardac 2050 (Polyquaternium-24 - Lonza	
Inc., Fair Lawn, NJ)	60.00%
Carsosoft CFI-90* (Lonza Inc.)	<u>33.30%</u>
Total	100.00%

## 10 **Procedure**

Carsosoft and Baradac ingredients were mixed together in a vessel. Next, water was added to the vessel and mixed. This was followed by the addition of HMHEC 3 while mixing. The pH of the mixture in the vessel was adjusted to 8.5 and mixed until all ingredients were dissolved. This formulation was then packed out in a container.

<sup>\*</sup> Carsosoft CFI-90: 1-Oleylamidoethyl-1-methyl-2-heptadecenylimidazolininon methosulfate

# Example 18 Automatic Dishwashing Detergent

	<u>Ingredients</u>	Weight %
	Sodium tripolyphosphate	39.50%
5	HMHEC 2	01.50%
	Makin NF-5 (Alkoxylated	
	isopropanolamide - Stepan)	03.00%
	Britesil H-20 (Sodium polysilicate -	
	PQ Corporation)	30.00%
10	CDB Clearon (Sodium	
	dichloroisocyanurate, Olin Corporation)	02.00%
	Sodium Carbonate, dense	<u>24.00%</u>
	Total	100.00%

## **Procedure**

Blender was charged with the carbonate ingredient. This was followed by the addition of the Makin ingredient and then the remaining ingredients. The blender was run for five minutes between each addition. The mixing was continued for 30 minutes or until homogeneously mixed. Once all the ingredients were added, the product was filled in a container.

## Example 19 Liquid Handsoap

	<u>Ingredients</u>	Weight %
5	Calblend clear (Sodium laureth sulfate (and) cocamide DEA (and) cocamidopropyl betaine - Pilot Chemicals,	
	Santa Fe Springs, CA)	30.00%
	HMHEC 1	00.50%
	Fragrance (strawberry)	00.25%
	Dye	00.05%
10	Water	69.20%
	Total	100.00%

## **Procedure**

HMHEC1 was added to the vortex of stirred water in a blender and was mixed until a slurry was formed. The pH of the slurry was adjusted to 8.5 and mixed for 45 minutes or until fully dissolved. Calblend clear ingredient was added next. This was followed by the addition of the fragrance and dye. The solution was mixed for 5 minutes between each addition and for 30 minutes after all of the ingredients were added.

# Example 20 Toilet Bar Soap

	<u>Phase I</u>	<u>Ingredients</u>	Weight %
	Α	Croamol CAP (Ceteary octanoate, Croda Inc., Parsippany, NJ)	10.00%
5	В	Promyristyl PM3 (PPG3 myristyl ether, Croda)	50.00%
	С	Procetyl AWS (PPG5-ceteth20, Croda)	08.00%
	Phase II		
	A	Sodium stearate	08.00%
Thomas and the state of the sta	В	HMHEC 2	02.00%
	Phase III		
STATE OF THE STATE	A	Deionized water	10.00%
Same of the same o	В	Propylene glycol	06.00% 06.00%
	С	Glycerin Total	100.00%

#### **Procedure**

First, ingredients in Phase I were blended together in a mixer. Next, they were heated to 80°C while continued mixing. Then, ingredients in phase II were added to the mixer and the temperature was maintained during the mixing. Once the ingredients in the mixer appeared to be homogeneous, ingredients of Phase III were added one at a time. The mixer was run on medium speed between each addition until the slurry appeared homogeneous. Next, the slurry was cooled to 65°C while mixing and then poured into a mold and packaged.

# Example 21 Toilet Bowl Cleaner

<u>Ingredients</u>	Weight %
Simulsol SL 10* (Seppic Co. Fairfield, NJ)	05.50%
Solagum SH210** (Seppic Co.)	01.50%
HMHEC 2	01.00%
Citric Acid	03.00%
Water	<u>89.00%</u>
Total	100.00%

## 10 **Procedure**

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Solagum and HMHEC 2 ingredients were added to the vortex of stirred water in a blender and then mixed for 45 minutes. This was followed by the addition of Simulsol and citric acid ingredients while mixing and then mixed for an additional 15 minutes and then packaged.

<sup>\*</sup> Simulsol SL10: Nonionic surfactant, glucose alkylpolyglucose

<sup>\*\*</sup> Solagum SH210: Aqueous solution of synthetic copolymer dispersed in isoparaffin

# Example 22 Toilet bowl Tablets, Rim Block, In-Cistrern Block

	<u>Ingredients</u>	Weight %
	Sodium sulfonate	49.99%
5	Ethoxylated fatty acid tallow	10.00%
	Tallow fatty acid monoethanolamide	03.00%
	HMHEC 1	09.00%
	Magnesium carbonate	04.00%
10	Sodium sulfate	20.00%
	Fragrance	04.00%
	Dye (FD&C blue 2 alum lake)	00.01%
	Total	100.00%

### **Procedure**

All of the ingredients, except the fragrance, were blended in a blender until homogeneous. The ingredients were added in the order listed above. The fragrance was sprayed into the blender while blending at medium speeds until homogeneous. The formulation was then extruded through a die at about 100°C and cooled to room temperature, cut, and packaged. The formulation could also contain other salts, such as sodium silicate, silicas, sodium carbonate, sodium citrate, etc. if needed to obtain desired pH, density, solubility etc.

# Example 23 Liquid Laundry Detergent

	<u>Ingredients</u>	Weight %
	Water	59.75%
5	HMHEC 3	00.25%
	Triethanol amine	02.00%
	Witco 1298* (Witco Dublin, OH)	05.00%
	propylene glycol	05.00%
	Witconol 25-7** (Witco)	15.00%
10	Varonic K205*** (Witco)	13.00%
-	Total	100.00%

## **Procedure**

HMHEC 3 was added to the vortex of stirred water in a blender, pH was adjusted to about 8.5, and mixed until dissolved. Next, triethanolamine was added to the blender and mixed for 30 minutes. The rest of the ingredients were added one at a time, in the order they are listed above, while mixing. Between each addition, the solution was mixed until the ingredients were dissolved. After all of the additions were made, the solution was mixed for an additional 30 minutes. The speed of agitation was adjusted as necessary to minimize foaming. The solution was packaged.

<sup>\*</sup> Witco 1298: Dodecylbenzene sulfonic acid linear

<sup>\*\*</sup> Witconol 25-7: Alcohol ethoxylate

<sup>\*\*\*</sup> Varonic K205: PEG-5 cocamine

# Example 24 Laundry Prespotter

	<u>Ingredients</u>	Weight %
	Water	77.10%
5	HMHEC 2	00.25%
	Witconol 25-7 (Witco)	07.00%
	Varonic K205 (Witco)	04.00%
	Witconate 45* liquid (Witco)	10.00%
	D'limonene	01.00%
10	Methyl paraben	00.15%
	Fragrance	<u>00.50%</u>
	Total	100.00%

## **Procedure**

The ingredients were added in the order listed above with constant agitation in a mixer. Mixing was continued until the mixture of the ingredients was lump free and homogeneous.

<sup>\*</sup> Witconate 45: Sodium dodecylbenzene sulfonate and sodium xylene sulfonate

# Example 25 Liquid Abrasive Cleaner

	<u>Ingredients</u>	Weight %
	Water	46.00%
5	Calcium carbonate	50.00%
	Acusol 820 (30% Acrylic polymer, Rohm and	
	Haas Company)	01.00%
	HMHEC 1	00.50%
	Bentonite clay (8.0% dispersion)	<u>02.50%</u>
10	Total	100.00%

## **Procedure**

The ingredients were added to the water in a mixer in the order that they are listed above. Next, pH was raised to 8.5 and the slurry was then mixed for 30 minutes or until lump free to form the final formulation.

# Example 26 Fabric Sizing Spray

	Phase I	<u>Ingredients</u>	Weight %
	Α	Deionized Water	68.35%
5	В	HMHEC 3	00.50%
	C	Carboxymethyl cellulose (CMC7L1T*, Hercules Incorporated	01.00%
	Phase II		
	Α	Sodium lauryl sulfate (Witcolate A, Witco)	00.05%
	В	Methylparaben	00.10%
10	C	Deionized Water	30.00%
		Total	100.00%

### **Procedure**

In Phase I, the polymers HMHEC 3 and CMC were added to the vortex of stirred water in a blender, the pH of the slurry in the blender was adjusted to 8.5, and the slurry was mixed until the polymers were fully dissolved. The surfactant and preservative were added to a separate mixer containing the water of Phase II and mixed until these components were fully dissolved. Phase II solution was then added to the vortex of Phase I solution and mixed for 10 minutes or until lump free. This formulation was then packed out in a container. For aerosol spray, the system was charged with a propellent.

<sup>\*</sup> CMC 7L1T: Sodium carboxymethylcellulose, with carboxymethyl substitution of 0.65 - 0.90 and 4% aqueous viscosity of 25-50 cps at 25°C.

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#### What is Claimed:

- 1. A household product composition comprising:
- (a) from about 0.1% to about 99% by weight of a vehicle system which comprises a hydrophobically modified water soluble polysaccharide polymer which comprises a water soluble polysaccharide polymer backbone, and a hydrophobic moiety selected from the group consisting of 1) 3-alkoxy-2-hydroxypropyl group wherein the alkyl moiety is a straight or branch chain having 3-24 carbon atoms, and 2) C<sub>3</sub>-C<sub>24</sub> alkyl, aryl alkyl, alkyl aryl groups and mixtures thereof, wherein the hydrophobic moiety is present in an amount up to the amount which renders said polysaccharide less than 1% by weight soluble in water and
- (b) at least one other household care ingredient.
- 2. The composition of claim 1 wherein the composition also comprises from about 0.01% to about 50% by weight of the household composition of a surfactant.
- 3. The composition of claim 2 wherein the surfactant is selected from the group consisting of anionic, nonionic, cationic, zwitterionic, and amphoteric, mixtures thereof.
- 4. The composition of claim 1 wherein the composition also comprises from about 0.1% to about 99% by weight of the household composition of a compatible solvent or solvent mixture.
- 5. The composition of claim 4 wherein the solvent or solvent mixture is selected from the group consisting of water, water-lower alkanols mixtures, polyhydric alcohols and mixtures thereof.
- 6. The composition of claim 5 wherein the solvent or solvent mixture is selected from the group consisting of water, propylene glycol, water-glycerine, sorbitol-water, water-ethanol, and mixtures thereof.

- 7. The composition of claim 2 wherein the composition also comprises from about 0.1% to about 99% by weight of the household composition of a compatible solvent or solvent mixture.
- 8. The composition of claim 1 wherein the hydrophobically modified polysaccharide backbone is selected from the group consisting of hydroxyethylcellulose (HEC), hydroxypropylcellulose (HPC), methylcellulose (MC), hydroxypropylmethylcellulose (HPMC), ethylhydroxyethylcellulose (EHEC), and methylhydroxyethylcellulose (MHEC), and agar, dextran, locust bean gum, starch, guar, and their nonionic derivatives, and mixtures thereof.
  - 9. The composition of claim 1 wherein the polysaccharide backbone is HEC.
- 10. The composition of claim 1 wherein the hydrophobic moiety is 3-butoxy-2-hydroxypropyl.
  - 11. The composition of claim 1 wherein the hydrophobic moiety is cetyl.
  - 12. The composition of claim 1 wherein the polysaccharide backbone is HPC.
- 13. The composition of claim 1 wherein the hydrophobic moiety is an alkyl radical having 4-16 carbons.
- 14. The composition of claim 4 wherein composition also contain an effective viscositying amount of a salt.
- 15. An air freshener/deodorizer composition comprising a solvent and an effective amount of the household composition of claim 4.
- 16. An insect repellent comprising an effective amount of the household composition of claim 4.

- 17. A bubble generating liquid comprising an effective amount of the household composition of claim 7.
- 18. A pet shampoo and shampoo-conditioner comprising an effective amount of the composition of claim 1.
- 19. A liquid soap product comprising an effective amount of the household composition of claim 7.
- 20. All purpose cleaner comprising an effective amount of household composition of claim 1.
- 21. A disinfectant comprising an effective amount of the household composition of claim 1.
- 22. A anti-itching lotion composition comprising an effective amount of the household composition of claim 4.
- 23. A bar soap composition comprising an effective amount of the household composition of claim 7.
- 24. A rug and upholstery cleaning composition comprising an effective amount of the household composition of claim 7.
- 25. A rug and upholstery composition comprising an effective amount of the household composition of claim 1.
- 26. A rug bleaching agent composition comprising an effective amount of the household composition of claim 1.

- 27. A laundry softener composition comprising an effective amount of the household composition of claim 7.
- 28. A dishwashing detergent composition comprising an effective amount of the household composition of claim 7.
- 29. A toilet bowl cleaner composition comprising an effective amount of the household composition of claim 7.
- 30. A laundry prespotter composition comprising an effective amount of the household composition of claim 1.
- 31. An auto cleaning composition comprising an effective amount of the household composition of claim 7.
- 32. The composition of claim 1 where the household composition also comprises an oil-in-water or water in oil emulsion.
- 33. A pet dental cleaning and deodorizing composition comprising an effective amount of the household composition of claim 1.

#### ABSTRACT OF THE INVENTION

A household preparation product composition is composed of from about 0.1% to about 99% by weight of a vehicle system which comprises a hydrophobically modified water soluble polysaccharide polymer which comprises a water soluble polysaccharide polymer backbone, a hydrophobic moiety of 1) 3-alkoxy-2-hydroxypropyl group wherein the alkyl moiety is a straight or branch chain having 3-24 carbon atoms, or 2) C<sub>3</sub>-C<sub>24</sub> alkyl, aryl alkyl, alkyl aryl groups and mixtures thereof, wherein the hydrophobic moiety is present in an amount up to the amount which renders said polysaccharide less than 1% by weight soluble in water, and at least one other household care ingredient. The composition can be used in a wide range of cleaning, polishing, sanitizing, pesticidal and toilet preparation applications such as air deodorants/fresheners, rug and upholstery shampoos, insect repellent lotions, children's bubble-making solutions, pet care products such as shampoos and deodorizer, soap bar and institutional liquid soap, all purpose kitchen cleaner and disinfectants, toilet bowl cleaners, fabric softener-detergent, fabric softener, fabric sizing agents, dishwashing detergents, vehicle cleaners and shampoos.

# COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; I believe I am the original, first and sole inventor of the subject matter which is claimed and for which a patent is sought on the invention entitled  Hydrophobically Modified Polysaccharides In Household Preparations the specification of which [x] is attached hereto [] was filed on and was amended on; I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above; and I acknowledge the duty to disclose information which is material to the examination of this application in accordance with title 37, Code of Federal Regulations, Sec. 1.56(a).					
Code, Sec. 119 of a certificate listed application for pat	I hereby claim foreign priority benefits under Title 35, United States Code, Sec. 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.				
Prior Foreign Appli	cation(s)		Priority Claimed		
(Number)	(Country)	(Day/Month/ Year Filed)	[] [] Yes No		
(Number)	(Country)	(Day/Month/ Year Filed)	[] [] Yes No		
I hereby claim the benefit under Title 35, United States Code, Sec. 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, Unites States Code, Sec. 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Sec. 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:					
(Application Seria	1 No.) (Fi	ling Date)	(Status)		
(Application Seria	l No.) (Fi	ling Date)	(Status)		

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: David Edwards, Reg. No. 27,293; Martin F. Sloan, Reg. No. 38,618; Mark D. Kuller, Reg. No. 31,925; Robert P. O'Flynn O'Brien, Reg. No. 35,629; Steven H. Markowitz, Reg. No. 27,095. Address all telephone calls to David Edwards at telephone no. (302) 594-6974. Address all correspondence to David Edwards, Hercules Incorporated, Intellectual Property Section, Hercules Plaza, Wilmington, Delaware 19894-0001.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Sec. 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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